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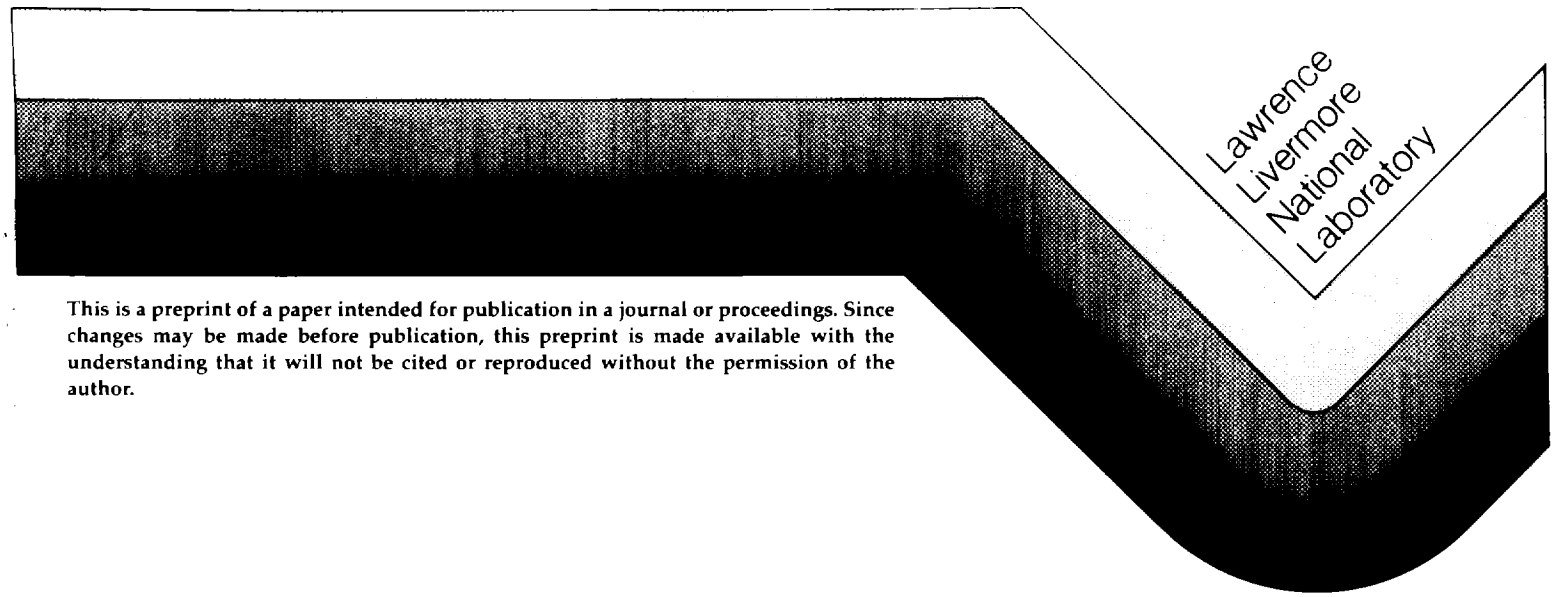

IRRADIATION DISORDERING AND ORDERING OF Cu_3Au BY FUSION NEUTRONS*

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Ordered and partially ordered Cu_3Au alloys (S = .30 - .99) have been irradiated at 4K and 300K with fusion neutrons at RTNS-II. The disordering rate was measured by monitoring electrical resistivity. The analysis of 4K irradiations and a comparison with fission reactor irradiations indicated that the disordering rate depended upon the long-range order parameter, S, $dS/d\phi t = -S(k_1 - k_2 S)$, where k_1 and k_2 are scaled with damage energy.

The results of 300K irradiation indicated that reordering competed significantly with disordering in the partially ordered sample. Compared to the results of 4K irradiation, the net disordering rate at 300K was higher than that at 4K. This difference and the dependence of disordering rate on S is discussed in terms of the effects of disorder and thermal displacements on cascade cooling processes.

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1. Introduction

A long range ordered alloy can be disordered or reordered by irradiation with energetic particles. The mechanisms of irradiation disordering and reordering had been reviewed in several papers [1,2]. Irradiation disordering has been attributed to the combination of several mechanisms, including (1) replacement collision sequences, (2) thermal spikes, (3) collapse of cascades to vacancy loops, (4) plastic spikes, and (5) random vacancy-interstitial recombination. A model for irradiation disordering has also been developed by Aronin [3], which predicts long range order parameter decreases exponentially with fluence. Agreement of experimental data with this model had been reported by several workers [3-7].

Irradiation reordering has been attributed to the diffusion of irradiation produced defects, and occurs at temperatures where defects are mobile. Irradiation reordering and disordering usually occur in competition with each other. A model describing the kinetics of ordering has been developed by Dienes [8]. Zee and Wilkes [9] applied this model, the rate theory of radiation produced defects [10] and the Aronin model [3] to explain the disordering and reordering of Cu_3Au at high temperatures. More neutron irradiation data are required to test these models.

In a previous paper [11], we reported the results of fusion neutron irradiation on Cu_3Au at 4.2K. It was found that the disordering rate could be scaled to fission neutron irradiation by damage energy cross section with a discrepancy of 20%. It was also found that the fusion

neutron disordering rate at 300K measured by the superlattice TEM technique [12], was a factor of two lower than our 4.2K data. There is therefore a further need to study fusion neutron irradiation on Cu_3Au to elucidate these differences. The objective of this paper is to study the effects of long range order and temperature on the neutron disordering rate of Cu_3Au.

2. Experimental procedures

2.1 Specimens

Fully ordered, partially ordered and fully disordered samples were supplied by Dr. M.A. Kirk of Argonne National Laboratory. All samples were fully annealed at high temperature. The fully ordered sample (S=1) was slowly cooled to 573K. The partially ordered sample (S=.77) was quenched from 655K to room temperature. The fully disordered sample was quenched from 873K, far above the order-disorder transition temperature, T_c = 663K.

Table 1 gives the electrical resistivities and long range order parameters, S, of these samples at 4.2K. The parameter S is calculated by Muto's equation [13], which relates resistivity to S in an ordered alloy. We note that the resistivity of samples was affected by how potential and current lead wires were attached. For the 300K irradiation samples, the wires were attached by solding with a $Pb_{40}Sn_{60}$ alloy of melting point 463K. The measured resistivities and calculated S for the fully ordered and partially ordered samples are consistent with values given by Kirk. On the other hand the disordered sample was partially reordered and the resistivity decreased.

For the 4.2K irradiation samples, the lead wires were attached by spot welding. The resistivities for the fully and partially ordered samples were increased and S decreased. Spot welding raised the sample temperatures above T_c and disordered the samples. Since this is a very fast process, we assumed that only the regions near the welds were affected and the central gage section was at the original state. This assumption results in a fraction of disordered (assumed S=0) gage section of .135 for the fully ordered sample and .188 for the partially ordered sample. In the data analysis, the resistance of this disordered portion was subtracted from the total resistance to reflect the resistivity behavior of the original material.

2.2 Fusion neutron irradiation

The 14.8 Mev fusion neutron irradiation was performed at the Rotating Target Neutron Source (RTNS-II) of Lawrence Livermore National Laboratory. Electrical resistivities of samples were measured in situ during irradiations with a four-point measurement technique. Typical sample gage dimensions were 10 mm in length and .38 mm in diameter. The potential lead wires had .13 mm diameter. The maximum error in resistivity due to uncertainty in gage dimensions was 1%.

The 4.2K experiment was conducted with a commercial Helitran cryostat. Niobium foils were spaced between resistivity samples to determine the final fluences of samples. Epoxy was used to electrically insulate samples and lead wires from the mounting copper substrate, dosimetry foils and other cryostat components. The details of the arrangement of samples, dosimetry foils, and lead wires were given previously [11].

The 300K experiment was conducted in a vacuum furnace maintained at an average pressure of 5×10^{-7} torr. Nb foils were spaced between samples to determine the final fluences. Samples, dosimetry foils and lead wires were mounted on a copper substrate (38 mm diameter 51 mm long), in which a Fire-Rod heater was embedded. Boron nitride coating was used to electrically insulate the samples and lead wires from the substrate and other furnance components. The sample temperature, monitored by two type S thermocouples, varied by 2K, and had an average of 299.14K. The measured resistivities were corrected to equivalent values at 300K by means of measured temperature coefficients, determined before and after irradiation. The differences in temperature coefficients before and after irradiation are less than 5%, and the average values were used. Prior to this experiment the samples were irradiated up to 433K to study reordering kinetics. These results will be presented elsewhere.

3. Results and discussions

3.1 Irradiation at 4.2K

The 4.2K irradiation results are summarized in Table 2 which lists resistivity, fluence, and S. An analysis of the data indicated that the resistivity change for the fully disordered sample could be fitted to the following relation,

$$\Delta\rho = \Delta\rho^{\infty} \left(1 - \exp\left(- \frac{\phi t}{\Delta\rho^{\infty}} \left(\frac{d\Delta\rho}{d\phi t}\right)_{\Delta\rho=0}\right)\right) \qquad (1)$$

where $\Delta\rho^{\infty}$ is equal to 5.89 nΩm and $\left(\frac{d\Delta\rho}{d\phi t}\right)_{\Delta\rho=0}$ is equal to $3.15\ X/0^{-22}$ nΩm^3/neutron. This equation reflects the resistivity damage due to defects. This amount of damage was subtracted from the

resistivity damage when calculating the changes in S for the fully ordered and partially ordered samples from Muto's equation [13].

3.2 Effect of S on disordering rate

Aronin [3] has shown that the irradiation disordering rate of an ordered alloy is given by $S = S_o \exp(-k \phi t)$, where S is the initial order parameter and k is a disordering constant independent of S, but dependent upon the type of irradiation. Using this equation, the disordering rate constants were calculated and given in Table 2. Most interesting about this result is that the constant k is not really independent of S, but increases with decreasing S.

3.3 Comparison with fission neutron disordering at 4.2K

The results of fission neutron irradiation at CP-5 facility by Kirk and Blewitt [14] are compared with our results. Their data are reproduced in Table 3, including the resistivity, order parameter at beginning, midway, and final damage, and the calculated disordering constant k. The order parameter was calculated in the same way as for our data. The resistivity increase due to defects was taken as that of the fusion neutron irradiation (Eq. 1) scaled by damage energy cross-section. The damage energy cross-sections used were 271 b-Kev [11] for 14 Mev neutrons, and 50.3 b-kev [11] for the CP-5 neutrons.

The results in Table 3 show that the disordering rate constant k decreases with S, in agreement with our fusion neutron irradiation results. The disordering rate constants for the CP-5 data were also scaled with damage energy cross-section to equivalent values for 14 Mev

neutrons and compared to our results in Fig. 1. Shown are also the data from our previous work [11] recalculated with the present value for defect resistivity (Eq. 1). The agreement between the two results are within 6.5%. Furthermore, it shows that k decreases with S linearly.

3.4 Irradiation results at 300K

The 300K irradiation results are summarized in Table 4 including resistivity, order parameter, fluence and disordering rate constant. The order parameter was calculated with Muto's equation [13]. In order to use this equation the resistivity at S=0 and at S=1, and the resistivity increase due to Frenkel pair defects need to be known. These values were determined as follows: The resistivities at 300K for samples of S=.9985 and S=.3012 were measured as 45.30 and 108.88 nΩm respectively. Using these data with Muto's equation, the 300K resistivity at S=1 and S=0 were calculated as 45.09 and 115.25 nΩm respectively. The defect resistivity increase at 300K was taken as the increase at 4.2K (eq. 1) multiplied by the fraction unrecovered after being isochronally annealed from 4.2K to 300K. This fraction was determined as 34% [15].

3.5 Effect of temperature on disordering rate

The disordering rate constants at 300K are compared to those at 4.2K in Fig. 1. Some significant observations are: (1) disordering rate at 300K is higher than at 4.2K by 56% at S=.985, and by 7.5% at S=.80; (2) the disordering rate at 300K is lower at S=.8 than at S=.98, indicating that reordering occurred in the former significantly.

For the sample at S=.3, the disordering rate appears to fall in line with dS/dt vs. S for the 4.2K data. However, this data point could be questionable. As mentioned in Section 2, this sample had been reordered during soldering. The soldering process was relatively slow, there could be a distribution of S in this sample from 0 at the center to some value, say .8, at the leads. Thus the measured disordering may only reflect the overall balance of reordering and disordering covering a wide range of S and it may be meaningless to assign this data to S=.3.

The measured disordering rate constant for the fully ordered sample is larger than the value of $8.5*10^{-24}$ m^2/n measured by the superlattice TEM technique, for RTNS neutrons at 300K [12]. The discrepancy we believe is due to the difficulty in measuring the volume of disordered zone by TEM. The contrast of the superlattice TEM image for a disordered zone is proportional to average value of S through the foil thickness. Combined with a minimum observable contrast it is possible that this underestimates the zone volume if S is much lower near the periphery of a cascade than at the core. A reanalysis of the TEM Data taking this into account, indeed showed that it was consistent with our data.

3.6 A possible mechanism for the effect of S and temperature on high energy neutron disordering

The increase of the disordering rate constant with increasing temperature and decreasing S has a simple interpretation with respect to the fundamental process of cascade disordering. At 300K, atoms are excited from their lattice positions by temperature, as a result, the

probability of long range energy focussing sequences is reduced. For the Cu_3Au structure, long-range focussing is along the [110] direction. In half these directions Cu atoms are aligned in rows. In the other half, Cu and Au alternate and focussing is limited due to the large mass difference. This low energy focussing provides an efficient mechanism for dissipating cascade energy. When the probability of this is reduced, the high defect density existing in the core of cascade is maintained at a higher temperature for a longer time. This leads to more defect motion and higher disordering rates in the cascade. A similar effect of thermal displacements on defect production by higher energy replacement sequences has been modeled by computer simulations in Cu [16]. The current work provides experimental evidence for this mechanism and further shows the possible consequence in a long-range ordered alloy.

The disruption of focussing sequences can also explain why the constant k increases with decreasing S. As S is decreased, the length of Cu chains is reduced, as a consequence, the number of long-range energy focussing sequences is reduced, leading to higher cascade temperature, more defect motion, and higher disordering rates.

3.7 Estimation of reordering rate at 300K

Based upon Dienes' model of reordering [8], the reordering rate for a fully ordered (S=1) sample can be neglected. It can be assumed that disordering at 300K is insensitive to S since focussing sequences have already been disrupted by the thermal displacement of atoms. Thus the reordering rate at S=.8 can be estimated as the difference

between the disordering rate at S=.985 and the disordering rate measured at S=.8, assuming there is no significant reordering at S=.985. This gives a value of 3.81×10^{-24} m^2/neutron for $dS/d\Phi t$.

Zee and Wilkes [9] had predicted the reordering rate of Cu_3Au at 302K by fast neutron irradiation based upon Kirk and Blewitt's CP-5 irradiation data [14] at higher temperature (400-423K). The predicted value for $dS/d\Phi t$ is 3.47×10^{-25} m^2/neutron. When scaled to a fusion neutron irradiation by damage energy cross section, the value is a factor of 2 lower than our data. Considering the critical dependence of their model on the vacancy ordering-jump energy, and the uncertainty of this energy, the agreement is reasonable. Further work is required to make a closer comparison.

4. Conclusions

We have studied fusion neutron irradiation disordering and reordering in Cu_3Au at 4.2 and 300K using electrical resistivity. The main results are summarized below:

1) Disordering rate at 4.2K can be expressed in the form of $dS/dt = -S*(k_1-k_2*S)$, where k_1 and k_2 are constants.
2) Both increasing temperature and decreasing S raise disordering rates. This is attributed to the disruption of low-energy focussed sequences leading to a slower cascade cooling rate.
3) Disordering by fusion neutrons can be scaled to that by fission neutrons using damage energy cross section ratios. Differences are within 6.5%.

5. Acknowledgements

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6. References

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Table 1. Summary of Initial Material Properties at 4.2K

Irra. Temp. (K)	4.2K Elec. Res (nΩm)	L. R. O. Parameter S
300	11.26	.9985
300	44.80	.7776
300	88.74	.3012
4.2	22.77	.9985*
4.2	54.54	.7776*
4.2	92.14	0

*Fractions of the gauge length of these two samples have been disordered during spot welding (see text).

Table 2. Summary of 4.2K Irradiation Results

S_o	S_f	$\Delta\rho$ ($n\Omega m$)	$\Delta\phi t$ ($10^{21} n/m^2$)	k ($10^{-24} m^2/n$)
.9985	.9534	8.31	2.726	16.94
.7776	.7126	9.49	4.270	20.41
0	0	1.18	4.182	--

Table 3. Results of CP-5 Irradiation at 4.2K for Cu_3Au

Sample	S_o	S_f	ρ_o (nΩm)	ρ_f (nΩm)	$\Delta\phi t$ $(10^{22}n/m^2)$	k $(10^{-24}m^2/n)$
Sample 1*	.9081	.8508	26.00	34.61	1.853	3.52
Sample 1**	.8508	.7887	34.61	43.31	2.040	3.72
Sample 2*	.8342	.7771	37.00	44.87	1.828	3.88
Sample 2**	.7771	.7148	44.87	52.81	2.066	4.04

*Data taken from beginning to middle of change.

**Data taken from middle to final change.

Table 4. Summary of 300K Irradiation Results

S_o	S_f	ρ_o (nΩm)	ρ_f (nΩm)	$\Delta\phi t$ (10^{20}n/m^2)	k (10^{-24}m^2/n)
.9885	.9790	46.683	48.036	3.666	26.40
.8055	.7986	69.718	70.543	4.009	21.64
.3006	.2971	108.904	109.095	4.100	28.74

*The small changes in S_o from Table 1 are due to changes in the samples during reordering experiments up to 433K prior to the 300K disordering experiments.

Figure Captions

Figure 1. RTNS-II damage-energy equivalent neutron disordering rate constant versus the degree of long-range order in Cu_3Au.

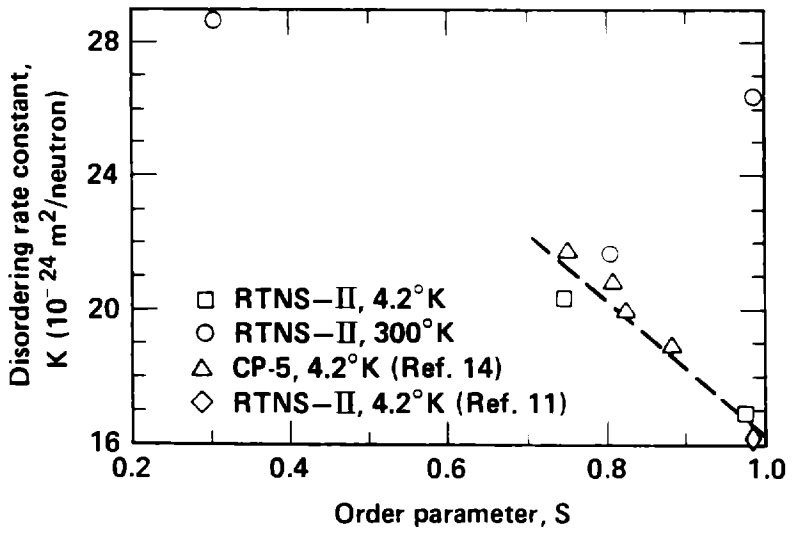
Disordering rate constant,
K (10^{-24} m^2/neutron)
28
24
20
16
0.2
0.4
0.6
0.8
1.0
Order parameter, S
RTNS–II, 4.2°K
RTNS–II, 300°K
CP-5, 4.2°K (Ref. 14)
RTNS–II, 4.2°K (Ref. 11)

Figure 1